First Principles Model of CANDU Fuel Phase Equilibrium

M.H. Kaye, C. Morrison, J.D. Higgs, F. Akbari, B.J. Lewis, and W.T. Thompson

Department of Chemistry and Chemical Engineering, Royal Military College of Canada,
Kingston, Ontario K7K 7B4

Abstract

Many modeling projects on nuclear fuel rest on a quantitative understanding of the co-existing phases at various stages of burnup. Since the various fission products have considerably different abilities to chemically associate with oxygen, and the O/M ratio is slowly changing as well, the chemical potential (generally expressed as an equivalent oxygen partial pressure) is a function of burnup. Concurrently, well recognized small fractions of new phases such as inert gas, noble metals, zirconates, etc. also develop. To further complicate matters, the dominant UO_2 fuel phase may be non-stoichiometric and most of minor phases have a variable composition dependent on temperature and possible contact with D_2O in the event of a sheathing defect.

The treatment discussed is a melding of several thermodynamic modeling projects over the past few years dealing with isolated aspects of this cornerstone multicomponent system. To simplify the computations, the number of elements has been limited to twenty major representative fission products known to appear in spent fuel. The proportion of elements was generated using ORIGEN except for oxygen, which was inferred from the concentration of the others. Provision to study the disposition of very minor fission products is included within the general treatment but these are introduced only on an as needed basis for a particular purpose. The building blocks of the model are the standard Gibbs energies of formation of the many possible compounds expressed as a function of temperature. To this data is added mixing terms associated with the appearance of the component species in particular phases. Sample output will be compared to the available direct knowledge gathered from post irradiation examination.

Introduction

The changes to fresh fuel as a result of burnup are illustrated as in Figure 1[1]. In general four phases are to be expected, namely the oxide matrix (the largest phase by far), noble metal inclusions, complex oxide inclusions, and a gas.

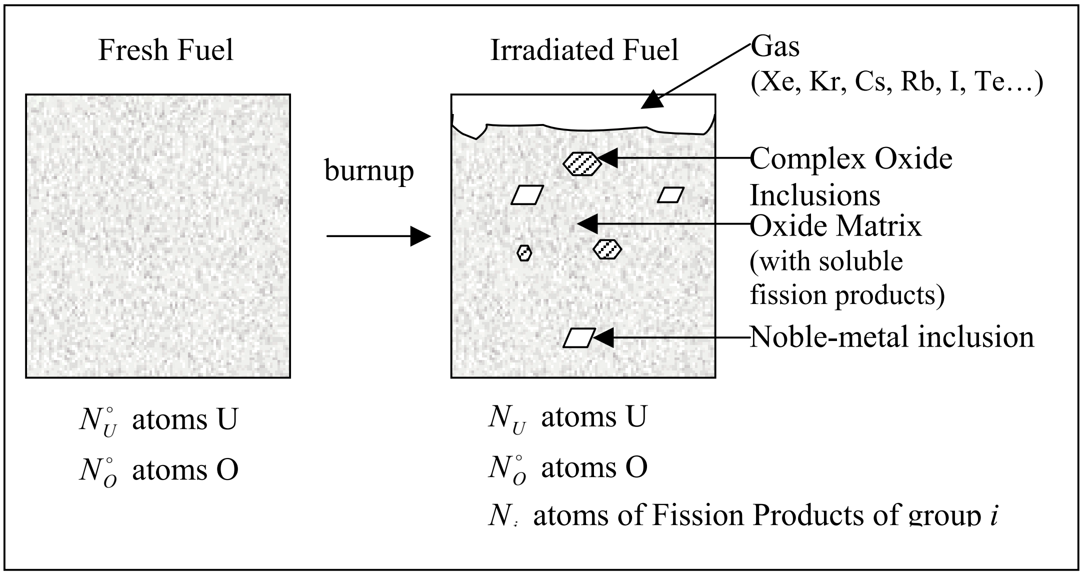

Figure 1. Typical result of irradiating fresh fuel, after Olander[1].

U-O Binary System

A thermodynamic model for the U-O binary system, shown in Figure 2, has been presented elsewhere[2-5], with comparisons to experimental data placing the phase boundaries. A principal feature of this model is that the deviation from ideal stoichiometric fluorite composition, UO_2, is accounted for by a hypothetical dissolved UO species or a hypothetical UO_3 species. The resultant relationship between temperature, O/U ratio in UO_{2+x} and partial oxygen pressure very closely follows Lindemer and Besmann[6].

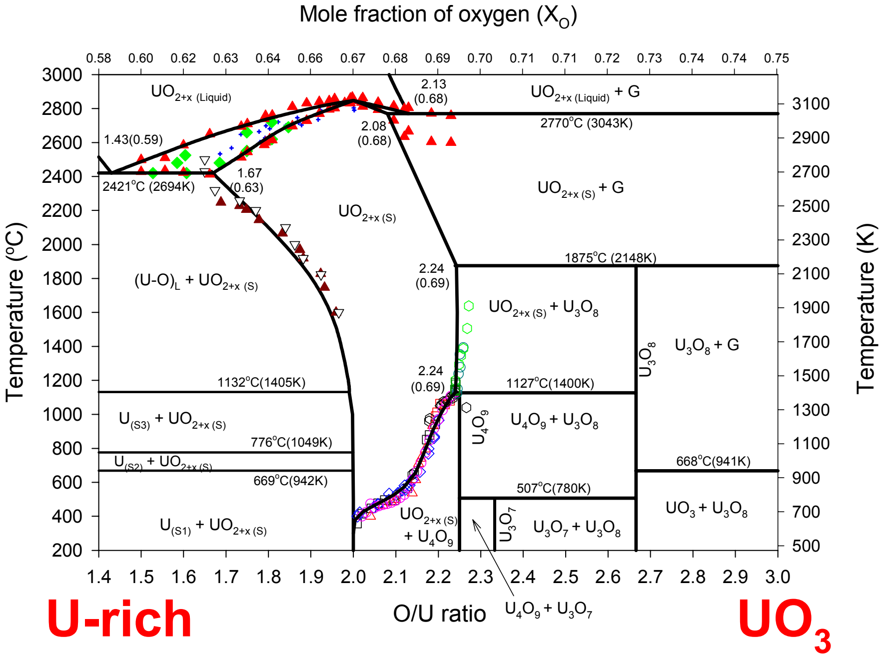

Figure 2. Computed U-O binary phase diagram based on Gibbs energy minimization compared to experimental data placing critical phase boundaries.

Solutes in UO_2

Experimental data for the solubility of oxides in UO_2 is difficult to obtain, especially at high temperatures. The current model incorporates 21 oxides, which are listed in Table 1.

Table 1. Oxides (grouped by formula, where M = metal) included as solutes in UO_2.

M_2O	MO	M_2O_3	MO_2
Cs_2O	SrO	Ce_2O_3	CeO_2
Rb_2O	BaO	Dy_2O_3	MoO_2
		Ho_2O_3	NbO_2
		La_2O_3	NpO_2
		Nd_2O_3	PuO_2
		Pr_2O_3	TeO_2
		Sm_2O_3	ZrO_2
		Y_2O_3	

In a review paper, Kleykamp[7] presents data for 12 of these oxides. In a typical experiment involving, for example Cs_2O, two samples of near stoichiometric powdered UO_2 (O/U = 2.003) were mixed with Cs_2O inside a thoria crucible. These crucibles were sealed within separate tantalum capsules, as shown in Figure 3. Each assembly was annealed at either 1273 or 2173K for 8 hours[7]. The experiment at the lower temperature did not attain thermodynamic equilibrium[7,8], but the results at 2173K yielded the following observations: the internal pressure of the Ta capsule was estimated, from the wall curvature, as 200 bars; an tantalum oxide layer deposited on the inner walls of the Ta capsule; and the level of dissolved Cs in the pellet was 0.07 mass% and 0.08 mol% Cs_2O, as determined by X–ray microanalysis.

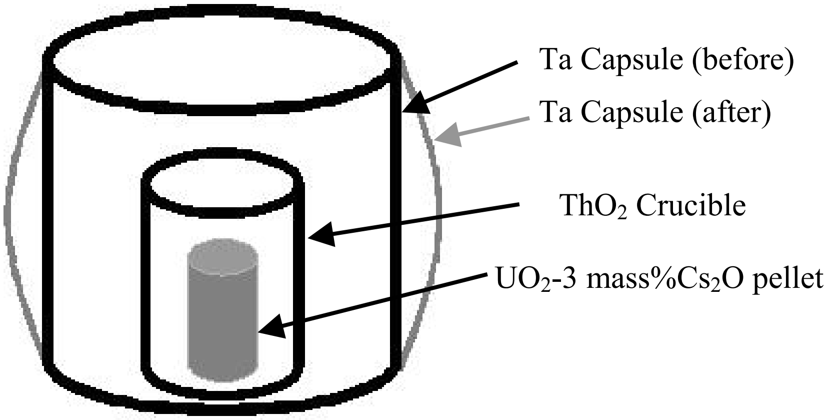

Figure 3. Schematic diagram of Cs_2O "Solubility" Experiment[7,8].

This experiment does not give the solubility of Cs_2O directly. Cs_2O dissociates, oxygen combines with Ta and a high Cs vapour pressure causes the capsule to swell. All of these factors must be considered in extracting the required thermodynamic data for Cs_2O dissolved in UO_2 solid solution. These data are often referred to as the lattice stability of the solute oxide in the structure of the solvent; that is:

$$Cs_2O_{(solid)} \rightarrow Cs_2O_{(infinite\ dilution\ in\ UO_{2\,(solid)})} \tag{1}$$

This lattice stability is the first entry in Table 2. Note that the number of moles of foreign solute cations per formula mass is also given. Solubility data along with the inferred lattice stabilities for selected lanthanide sequioxides[7] also appear in Table 2.

Table 2. Thermodynamic data for selected solute oxides in UO_2.

Compound	Solubility in UO_2 (%)	Temperature (K)	Lattice Stability (w.r.t. solid) (J/mol)	Cations per Formula mass
Cs_2O		2173	230	2
La_2O_3	69	523	-2686+8.505T	2
La_2O_3	50	1523		2
Pr_2O_3	55	1523	-4268+8.505T	2
Nd_2O_3	68	1523	-7714+8.505T	2
PuO_2	totally miscible		0	1
NpO_2	totally miscible		0	1

The behaviour adopted for PuO_2 was derived from the PuO_2-UO_2 diagram, which was determined by Lyon and Baily[9]. Their results, together with the calculated phase diagram, are shown in Figure 4. This diagram, based on ideal mixing of the actinide oxides in UO_2, shows that UO_2 and PuO_2 are completely miscible in both the solid and liquid phases, which is confirmed elsewhere[10,11].

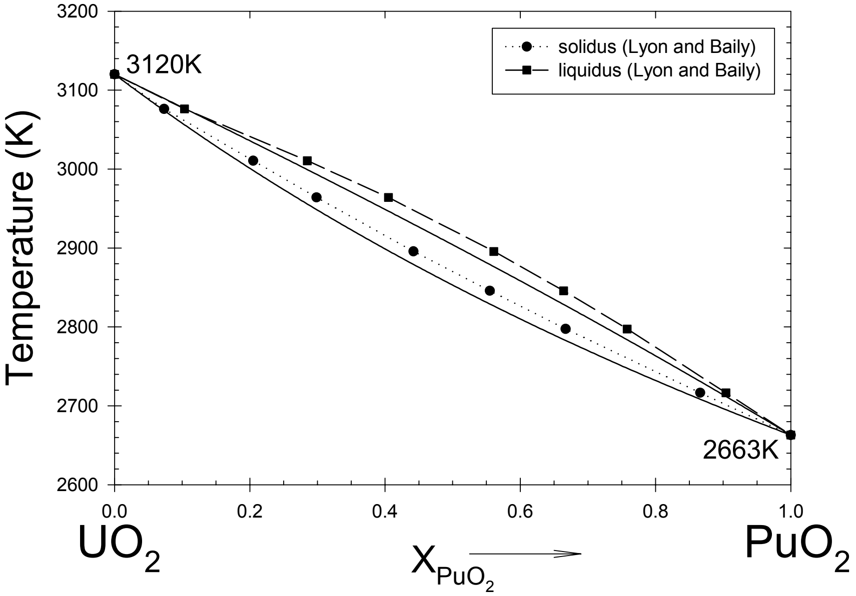

Figure 4. UO_2 – PuO_2 phase diagram determined by Lyon and Baily.

In general the agreement shown here is sufficient, considering the lack of data for this system and the error inherent for measurements at extremely high temperatures, to make this model acceptable.

A similar treatment was applied for the UO_2–NpO_2 binary system, using melting data taken from Russian work[12,13]. The lattice stabilities for PuO_2 and NpO_2 in Table 2 are set to zero. For the UO_2–ZrO_2 binary system, a complete model incorporating the three crystal structures of ZrO_2 has been developed[5].

Noble Metals

The presence of noble metal inclusions (the so-called “white” or “five metal” inclusions) in irradiated fuel has been well documented[7,14-20] and thermodynamic models for these inclusions have been developed, validated, and presented previously[21-23]. A phase diagram representative of the Mo-Pd-Rh-Ru quaternary is shown in Figure 5. This may be understood as four Gibbs triangles for the ternary combinations of elements. The triangles are arranged so that Rh-Pd, Rh-Ru, and Pd-Ru binary edges on the central triangle are common to the surrounding three ternary systems.

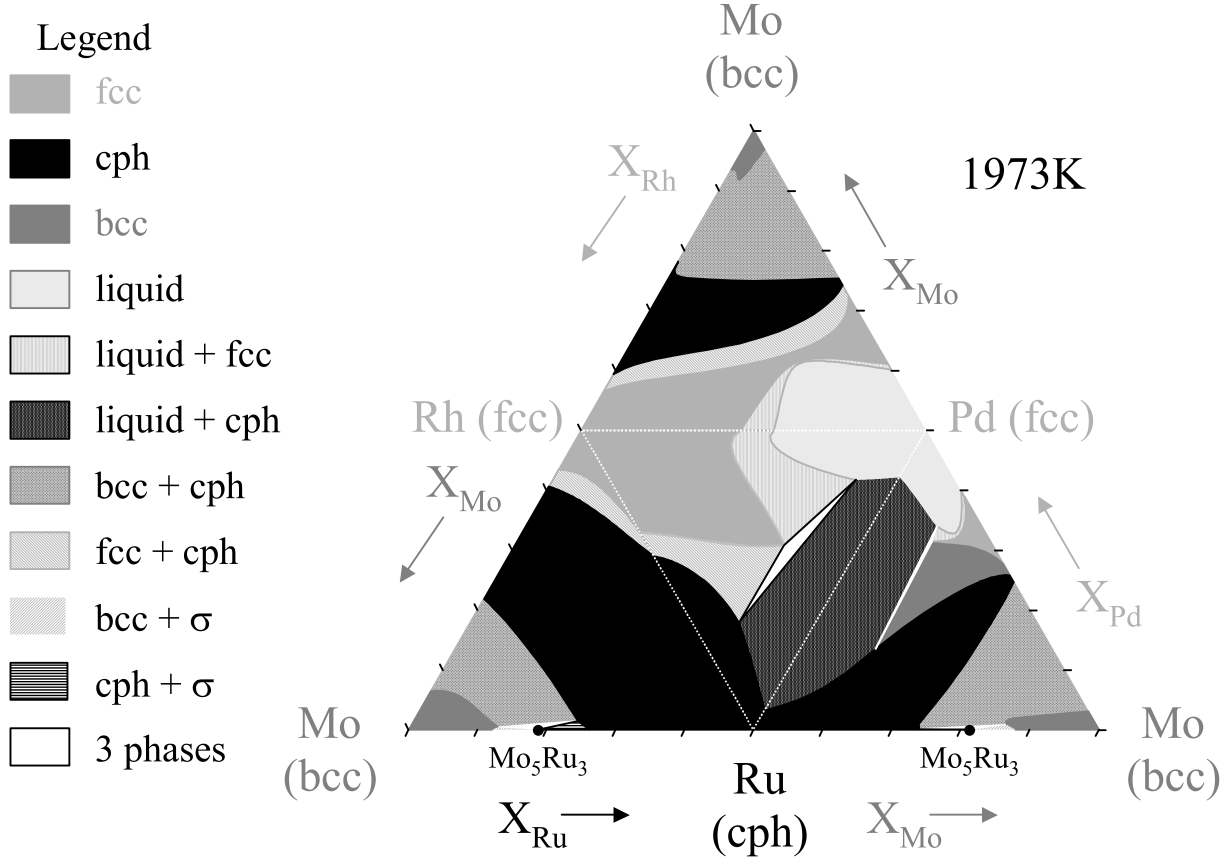

Figure 5. Noble Metal Phase diagram showing the faces of a representative tetrahedron unfolded on to a plane.

In the results of a typical calculation shown later in Figure 7, the metallic inclusions are in the close packed hexagonal crystal structure, which is to be expected[16,24]. Furthermore, it has been reported[16] that for alloys with low Mo content in the alloy (e.g., when the Mo is depleted by the presence of excess oxygen), the fcc-solid phase appears in conjunction with the cph-solid phase. The noble metal inclusions may be two phased under some circumstances. The present treatment accounts for this possibility.

Non-Fluorite Oxide Phases

A separate oxide phase is known to occur in irradiated fuel and is well documented[1,17,19,25]. These oxide inclusions of the type $(Ba,Sr)(U,Pu,Zr,Ln,Mo)O_3$, form a "grey" phase with a perovskite structure[19], and result because of the extremely limited solubility of BaO and SrO in other actinide oxides[1]. However, the presence of zirconium generally leads to the formation of zirconates. As a result of X-ray microanalysis of this grey phase, Kleykamp includes Cs with the Ba and Sr[7], which agrees with others[17]. Thus, it is not surprising to see the formation of one of these compounds in a calculation, and in fact, others form as well when the level of oxygen in the system increases.

Given the lack of data, the current treatment of the four distinct solid oxide phases and one liquid oxide phase, listed below, is to assume ideal solution behaviour. The ideal mixing treatments, it should be noted, recognize the number of moles of ions contributed to the solid solution in relation to the formula mass of the oxide components.

i. (Rb/Cs) Zirconate solid – $(Rb,Cs)_2ZrO_3$
ii. (Sr/Ba) Zirconate solid - $(Sr,Ba)ZrO_3$
iii. (Rb/Cs) Uranate solid - $(Rb,Cs)_2UO_4$
iv. (Sr/Ba) Uranate solid - $(Sr,Ba)UO_4$
v. (Rb/Cs) Molybdenate liquid – $(Rb,Cs)MoO_4$

Of course the appearance of Cs, for example, in combination with Mo, Zr, or U in condensed phases, does not preclude Cs also existing in the vapour phase or as a dissolved oxide in UO_{2+x}.

The "Other" Metallic Phase (UPd_3-URh_3-URu_3

As noted by Imoto[25], post-irradiation examination[26,27] of fast-breeder reactor (FBR) fuels has shown the presence of $(U,Pu)Pd_3$ particles. It should be noted that similar metallic inclusions of URu_3 and URh_3 may also be possible[28,29]. In the current treatment this phase is considered to be an ideal solution of the three solids UPd_3, URh_3, and URu_3, data for which was derived from Cordfunke and Konings[30]. Again the ideal mixing recognizes three atoms per mole of each component in the formulation.

Rhombohedral Oxide Phase

A potentially important solution phase involving compounds of the stoichiometry $UO_3{\cdot}(Ln_2O_3)_3$, where Ln may be any lanthanide, has been recently been added to the model[31]. This phase, treated as an ideal solution, has a rhombohedral structure and includes all the lanthanides except Pm, Tm, and Yb. Generally speaking this phase does not appear in normal fuel, but the nearness to precipitation (i.e., the activity) is output from the model.

Establishing the Fuel Inventory for Equilibrium Calculations

Fuel Inventory

The fission product inventory for any particular fuel, at a given burn-up, was generated by the ORIGEN 2 code. For a particular burnup, the code provides the inventory for the isotopes of approximately 63 elements. The greater part of these are present in very small amounts, so our calculations are only concerned with the 20 most important elements, ranked both by relative concentration (e.g., Pu, Zr, and Mo) and radiological consequence (e.g., I, Cs, Xe, and Ru). In addition O_2 and H_2 are added to the calculation to allow for the possible presence of steam in the event of a sheathing breach.

A typical CANDU fuel composition after 175MWh/kgU burnup[1] is given in Figure 6. In the calculation, the Xe accounts for the inventory of all inert gases; mols of Kr are combined with Xe. It is also important to determine the correct oxygen inventory for the fuel equilibria calculation. For a given fuel burnup, the amount of available oxygen was determined on the premise that the original mass of fuel was only stoichiometric UO_2.

79.95107 **U** + 0.29738 **Pu** + 0.16719 **Zr** + 0.19846 **Xe** +
0.12592 **Mo** + 0.10593 **Ce** + 0.11598 **Ru** + 0.09257 **Nd** +
0.05189 **Sr** + 0.08325 **Cs** + 0.01607 **Np** + 0.04518 **Ba** +
0.03903 **La** + 0.02548 **Y** + 0.03739 **Tc** + 0.02832 **Pr** +
0.01635 **Rb** + 0.01744 **Te** + 0.04029 **Pd** + 0.00971 **I** +
0.01653 **Rh** + 0.00009 **H** + 80.85685 $\mathbf{O_2}$ + x $\mathbf{H_2O}$=

Figure 6. The inventory of a CANDU fuel bundle at 175MWh/kgU burnup.

Computational Methodology

The calculation of the fuel phase equilibrium (as distinct from the gross proportion of atoms) involves distributing oxygen among the candidate elements and calculating the Gibbs energy change from an initial condition where the elements are imagined to be uncombined chemically. Successive redistributions are made in a systematic way[32] until the Gibbs energy change can be made no more negative. The calculations depend mainly on the Gibbs energy of formation of the various oxides. However there are other contributions to the computation of Gibbs energy change that are associated with the existence of species in different phases. For the gas phase, it is sufficient (given the uncertainties of the data) to correct for the concentration of the component molecules or atoms by adding to the standard Gibbs energy

$$\overline{G} - G^{\circ} = \Delta G = RT \ln\left[XP\right] \tag{2}$$

where $\overline{G}$ is the molar Gibbs energy of the dissolved component; G^o is the molar Gibbs energy of the pure gas at 1 atm pressure; R is the gas constant; T is the absolute temperature; X is the atom or mole fraction of the particular gaseous component; and P is the total pressure. For the condensed solution phases, the correction to the Gibbs energy for concentration is similar but

---

[1] Bin#10, power 980.3kW(f)/bundle, irradiated for 143 days, burnup 140 MWdays, and flux $=2.16\times10^{14}$N/cm$^2\cdot$sec

involves the activity coefficient as well as the mole fraction of that component in the phase involved.

$$\overline{G} - G^{\circ} = \Delta G = RT \ln\left[\gamma X\right] \qquad \textbf{(3)}$$

where $\overline{G}$ is the molar Gibbs energy of the dissolved component; G^{o} is the molar Gibbs energy of the pure condensed phase, which is not sensitive to the hydrostatic pressure in the range of interest (0-100 atm); and γ is the activity coefficient.

In dilute solution, such as the fission products in UO_2, γ is nearly concentration independent but temperature dependent where the data warrant. Care must be taken in calculating the value of X for each component particularly in UO_2 phase, since the formula mass for the component oxide may be associated with the introduction of more than one mole of cations (i.e., “foreign species”) to the solution. For example, one mole of La_2O_3 dissolved in UO_2 contributes 2 moles of La^{3+}; this factor must be taken into account in calculating concentrations for the UO_2 phase. There is relatively little data to guide the choice of activity coefficients for each oxide component other than the findings gathered by Kleykamp[7,33].

A Typical Burn-up Calculation

The results of a typical burnup calculation are presented in Figure 7.

The instigation to develop a First Principles Fuel model was in support of the Industry Standard Toolset (IST) SOURCE 2.0 code[21,34,35]. In the original treatment, inspired by Cubicciotti[36-39], the Gibbs energy minimization calculations were based on the assumptions[35] that: all gases mix ideally; the liquid metallic elements, if present, mix in an ideal solution; and all other condensed species are mutually insoluble stoichiometric compounds.

The main oversimplification was the non-stoichiometry in the UO_{2+x} phase and the lack of non-ideal solution behaviour of both the solute oxides in the fuel phase and the elements in the noble metal phase. The results captured in SOURCE 2.0 by the Method of Chemical Potentials[21,34,35], differ significantly from the present treatment which addresses in greater detail the known aspects of the phase equilibrium.

Mole fraction precedes each component

Relative mols of the phases

```
                      +  0.21849           Cs
                      +  0.41384E-01       Rb
                      +  0.31815E-01       CsI
                      +  0.62750E-02       Cs2MoO4
                      +  0.22883E-02       RbI
                      +  0.14364E-02       Rb2MoO4
                      +  0.31636E-03       MoOH
                      +  0.13332E-03       Cs2
                      +  0.96285E-04       Pd
                      +  0.58623E-04       BaO
                      +  0.49708E-04       RbCs
                      +  0.11919E-04       Cs2I2
                      +  0.26367E-07       O2
...
                     (2000 K, 1 atm, gas)
+ 80.820 mol          (  0.98878           UO2
                      +  0.36795E-02       PuO2
                      +  0.20687E-02       ZrO2
                      +  0.14960E-02       MoO2
                      +  0.64204E-03       SrO
                      +  0.57269E-03       Nd2O3
                      +  0.55881E-03       BaO
                      +  0.47468E-03       Ce2O3
                      +  0.36132E-03       CeO2
                      +  0.32762E-03       UO3
                      +  0.24146E-03       La2O3
                      +  0.21579E-03       TeO2
                      +  0.19884E-03       NpO2
                      +  0.17520E-03       Pr2O3
                      +  0.15763E-03       Y2O3
                      +  0.35992E-04       Cs2O
                      +  0.12488E-04       Rb2O
                      +  0.11018E-06       UO
                     (2000 K, 1 atm, Fluorite UO2+x with dissolved oxides)
+ 0.17869 mol         (  0.64871           Ru
                      +  0.20924           Tc
                      +  0.92015E-01       Rh
                      +  0.44908E-01       Pd
                      +  0.51248E-02       Mo
                     (2000 K, 1 atm, hP2 - Noble Metal Phase)
+ 0.10795E-01 mol     (  0.99545           UPd3
                      +  0.27033E-02       URh3
                      +  0.18451E-02       URu3
                     (2000 K, 1 atm, Other Metallic Phase - U(Pd,Rh,Ru)3)
+ 0.18135E-02 mol     (  0.70420           Cs2MoO4
                      +  0.29580           Rb2MoO4
                     (2000 K, 1 atm, Liquid solution of (Cs/Rb)MoO4)
```

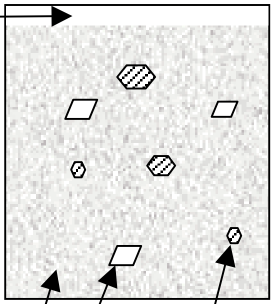

Figure 7. The results of a typical burnup calculation (CANDU 175 MWh/kgU) based on the atomic proportions shown in Figure 1. Here the reactants may form solutions (described previously), as well as separate condensed phases. Other phases are involved in the computation, but do not appear at this burnup.

Conclusions

A multi-component model involving the major fission products has been developed that can account for the following well-documented aspects of nuclear fuel chemistry:

a. non-stoichiometry in the UO_2 fluorite phase;
b. dilute solution behaviour of significant solute oxides;
c. noble metal inclusions of various potential phases;
d. zirconate and uranate solutions;
e. a molybdenate solution;
f. a second metal solid solution $U(Pd\text{-}Rh\text{-}Ru)_3$; and
g. volatile species within the inert gas phase.

The current treatment allows for direct use in a variety of applications. For example, a loss-of-coolant accident in which the fuel is exposed to a changing atmosphere of hydrogen and steam at elevated temperature. At the other end of the temperature spectrum, calculations have been applied involving aqueous media yielding the leaching of fission products from the fuel of defected elements. The same considerations surround the disposal of spent fuel. Currently adjustments, in an ongoing project, are being applied to allow application to Dy-doped fuel.

Acknowledgements

The authors thank the CANDU Owners Group, Atomic Energy of Canada Limited, the Director General for Nuclear Safety, and the Natural Sciences and Engineering Research Council of Canada for their support.

---

1. D.R. Olander, “Chapter 12- Behaviour of Solid Fission Products in Oxide Fuel Elements”, Fundamental Aspects of Nuclear Reactor Fuel Elements, (1976), 172-198.
2. B.J. Lewis, W.T. Thompson, F. Akbari, D.M. Thompson, C. Thurgood, and J.D. Higgs, “Thermodynamic and Kinetic Modelling of Fuel Oxidation Behaviour in Operating Defective Fuel”, *Journal of Nuclear Materials*, **328** (2004), 180-196.
3. B.J. Lewis, W.T. Thompson, J.D. Higgs, F. Akbari, M.H. Kaye, C. Thurgood, J. Serdula, and C. Morrison, “Modelling of Fuel Oxidation Behaviour in Defective Fuel”, COG Report - COG-04-2043, (March 2005).
4. J.D. Higgs, B.J. Lewis, W.T. Thompson, and Z. He, “A Conceptual Model for the Fuel Oxidation of Defective Fuel”, 9th International Conference on CANDU Fuel: Belleville ON, (2005).
5. F. Akbari, M.J. Welland, B.J. Lewis, and W.T. Thompson, “Considerations in Modelling the Melting of Fuel Containing Fission Products and Solute Oxides”, 9th International Conference on CANDU Fuel: Belleville ON, (2005).
6. T.B. Lindemer and T.M. Besmann, “Chemical Thermodynamic Representation of $UO_{2\pm x}$,” *Journal of Nuclear Materials*, **130** (1985), 473-488.

---

7. H. Kleykamp, "The Solubility of Selected Fission Products in UO_2 and $(U,Pu)O_2$,", *Journal of Nuclear Materials*, **206** (1993), 82-86.

8. H. Kleykamp, "The Chemical State of the Fission Products in Oxide Fuels", *Journal of Nuclear Materials*, **131** (1985), 221-246.

9. W.L. Lyon and W.E. Baily, "The Solid-Liquid Phase Diagram for the UO_2-PuO_2 System", *Journal of Nuclear Material*, **22** [3] (1967), 332-339.

10. H. Kleykamp, "Phase Equilibria in the UO_2-PuO_2 System Under a Temperature Gradient", *Journal of Nuclear Materials*, **294** (2001), 8-12.

11. T. Philipponeau, "Melting Point of U-Pu Mixed Oxide Fuels", Fast Reactor European Collaboration Data Sheet, (1990).

12. Yu. I. Belyaev, *Radiokhimiya*, **25** [6] (1983), 791-794.

13. T.D. Chikalla, C.E. McNeilly, J.L. Bates, and J.J. Rasmussen, International Colloquium on the Study of Crystalline Transformation at High Temperature Above 2000 K, Odeillo, France, (1971).

14. H. Kleykamp, J.O.A. Paschoal, R. Pejsa, and F. Thümmler, "Composition and Structure of Fission Product Precipitates in Irradiated Oxide Fuels: Correlation with Phase Studies in the Mo-Ru-Rh-Pd and BaO-UO_2-ZrO_2-MoO_2 Systems" *Journal of Nuclear Materials*, **130** (1985), 426-433.

15. H. Kleykamp, "The Chemical State of the Fission Products in Oxide Fuels within the Different Stages of the Nuclear Fuel Cycle", High Tech Ceramics, edited by: P. Vincenzini, Elsevier Science Publishers: Amsterdam, (1987), 2769-2798.

16. H. Kleykamp, "The Chemical State of Fission Products in Oxide Fuels at Different Stages of the Nuclear Fuel Cycle", *Nuclear Technology*, **80** (1988), 412-422.

17. T. Muromara, T. Adachi, H. Takeishi, Z. Yoshida, T. Yamamoto, and K. Ueno, "Metallic Phases Precipitated in UO_2 Fuel- I. Phases in Simulated Fuel", *Journal of Nuclear Materials*, **151** (1988), 318-326.

18. S.G. Prussin, D.R. Olander, W.K. Lau, and L. Hansson, "Release of Fission Products (Xe, I, Te, Cs, Mo, and Tc) from Polycrystalline UO_2", *Journal of Nuclear Materials*, **154** (1988), 25-37.

19. P.G. Lucuta, R.A. Verrall, Hj. Matzke, and B.J. Palmer, "Microstructural Features of SIMFUEL- Simulated High-Burn-up UO_2-based Nuclear Fuel", *Journal of Nuclear Materials*, **178** (1991), 48-60.

20. S. Yamanaka and K. Kurosaki, "Thermophysical Properties of Mo-Ru-Rh-Pd Alloys", *Journal of Alloys and Compounds*, **353** (2003), 269-273.

21. M.H. Kaye, A Thermodynamic Model for Noble Metal Alloy Inclusions in Nuclear Fuel Rods and Application to the Study of Loss-of-Coolant Accidents, *Doctoral Thesis*, Queen's University, (2001).

22. M.H. Kaye, W.T. Thompson, B.J. Lewis, S. Sunder, and R. O'Connor, "A Thermodynamic Model of the Mo-Pd-Rh-Ru-Tc Quinary System: Metallic Inclusions in

---

Nuclear Fuel Rods', 10th International Symposium on Thermodynamics of Nuclear Materials (STNM-10): Halifax, Nova Scotia, (August 2000).

23. M.H. Kaye, W.T. Thompson, B.J. Lewis, S. Sunder, and R. O'Connor, "Thermodynamic Properties of Noble Metal Inclusions in Nuclear Fuel Rods", Proceedings of the 7th International Conference on CANDU Fuel: Kingston ON, (September 2001).
24. H. Kleykamp, "The Chemical State of LWR High Power Rods Under Irradiation", *Journal of Nuclear Materials*, **84** (1979), 109.
25. S. Imoto, "Chemical State of Fission Products in Irradiated UO_2", *Journal of Nuclear Material*, **140** (1986), 19-27.
26. F.T. Ewart, R.G. Taylor, J.M. Horspool, and G. James, "The Chemical Effects of Composition Changes in Irradiated Oxide Fuel Materials II-Fission Product Segregation and Chemical Equilibria", *Journal of Nuclear Material*, **61** (1976), 254-270.
27. T. Itaki, K. Kono, H. Tachi, S. Yamonouchi, S. Yuhara, and I Shibahara, *Journal of Atomic Energy Society of Japan*, **27** (1985), 435-449.
28. M.S. Chandrasekhariah, "Oxygen Potential and the Chemical State of the Fission Products Ruthenium, Rhodium, and Palladium in Irradiated Oxide Fuels", *Journal of Nuclear Material*, **130** (1985), 366-374.
29. K. Kurosaki and M. Uno, "Phase Equilibria in the Ternary URu_3-URh_3-UPd_3 System", *Journal of Alloys and Compounds*, **271-273** (1998), 641-644.
30. E.H.P. Cordfunke and R.J.M. Konings, Thermochemical Data for Reactor Materials and Fission Products, Elsevier Science Publishers: Amsterdam, (1990).
31. S. White, S. Vogel, M.H. Kaye, B.J. Lewis, and W.T. Thompson, "Thermodynamic Modelling of the U-Dy-O System", 9th International Conference on CANDU Fuel: Belleville ON, (2005).
32. C.W. Bale, A.D. Pelton, and W.T. Thompson. "Facility for the Analysis of Chemical Thermodynamics - User Manual 2.1", Ecole Polytechnique de Montreal/ McGill University, (1996).
33. C.W. Morrison, F. Akbari, and W.T. Thompson, "Properties of Selected Oxide Fission Products in UO_2", RMC Nuclear Engineering Group - Internal Report, (March 4, 2004).
34. B.J. Corse, "FORM 2.0- Fuel Oxidation and Release Model- A Computer Code to Predict the Low Volatile Fission-Product Release and Fuel Volatilization from Uranium Dioxide Fuel Under Severe Reactor Accident Conditions", *Master's Thesis*, The Royal Military College, (1997).
35. B.J. Lewis, B.J. Corse, W.T. Thompson, M.H. Kaye, F.C. Iglesias, P. Elder, R. Dickson, and Z. Liu, "Low Volatile Fission Product Release and Fuel Volatilization During Severe Reactor Accident Conditions", *Journal of Nuclear Materials*, **252** (1998), 235-256.
36. D. Cubicciotti, "A Model for Release of Fission Gases and Volatile Fission Products from Irradiated UO_2 in Steam Environment", *Nuclear Technology*, **53** (1981), 5-7.
37. D. Cubicciotti and B.R. Sehgal, "Vapour Transport of Fission Products in Postulated Severe Light Water Reactor Accidents", *Nuclear Technology*, **65** [5] (1984), 266-291.

38. D. Cubicciotti, "Vaporization Thermodynamics of Fission Products from Fuel under Nuclear-Accident Conditions", *Advances in Ceramics*, **17** (1986), 211-222.

39. D. Cubicciotti, "Vapour Transport of Fission Products under Nuclear-Accident Conditions", *Journal of Nuclear Materials*, **154** (1988), 53-61.